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Sub-gap optical response in the Kitaev spin-liquid candidate -RuCl3

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Abstract

© 2018 IOP Publishing Ltd. We report detailed optical experiments on the layered compound -RuCl3 focusing on the THz and sub-gap optical response across the structural phase transition from the monoclinic high-temperature to the rhombohedral low-temperature structure, where the stacking sequence of the molecular layers is changed. This type of phase transition is characteristic for a variety of tri-halides crystallizing in a layered honeycomb-type structure and so far is unique, as the low-temperature phase exhibits the higher symmetry. One motivation is to unravel the microscopic nature of THz and spin-orbital excitations via a study of temperature and symmetry-induced changes. The optical studies are complemented by thermal expansion experiments. We document a number of highly unusual findings: A characteristic two-step hysteresis of the structural phase transition, accompanied by a dramatic change of the reflectivity. A complex dielectric loss spectrum in the THz regime, which could indicate remnants of Kitaev physics. Orbital excitations, which cannot be explained based on recent models, and an electronic excitation, which appears in a narrow temperature range just across the structural phase transition. Despite significant symmetry changes across the monoclinic to rhombohedral phase transition and a change of the stacking sequence, phonon eigenfrequencies and the majority of spin-orbital excitations are not strongly influenced. Obviously, the symmetry of a single molecular layer determines the eigenfrequencies of most of these excitations. Only one mode at THz frequencies, which becomes suppressed in the high-temperature monoclinic phase and one phonon mode experience changes in symmetry and stacking. Finally, from this combined terahertz, far- and mid-infrared study we try to shed some light on the so far unsolved low energy (<1 eV) electronic structure of the ruthenium 4d 5 electrons in -RuCl3.

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Keywords

dielectric and optical properties, FIR and MIR spectroscopy, Kitaev spin liquids, orbital excitations, phonons, structural phase transition, THz, α-RuCl 3

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